Title: **HIGH EFFICIENCY DESULFURIZATION OF SYNTHESIS GAS: II**

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Subcontractor: None
Industrial Collaborator: None
Grant Number: DE-FG-00NT40813
Performance Period: September 2000 – August 2003

ABSTRACT

A previous study has shown that reduced cerium oxide, CeO_n with $1.5 < n < 2,0$, was capable of reducing the H_2S concentration to 1 ppmv or less at temperatures in the range of 650^{o}C to 800^{o}C. This is a considerable improvement over the performance of current generation zinc-based sorbents at these conditions, and shows promise for achieving Vision 21goals of reducing the H_2S in coal gasification process streams to sub-ppmv levels. In addition, the sulfided product, Ce_2O_2S, can be easily regenerated with the direct formation of elemental sulfur, thereby avoiding the need for further treatment of the regeneration product gas to prevent atmospheric emissions of SO_2.

Because of the stability of CeO_2, an oxygen-free, high H_2 gas was required to achieve reduction. Gases produced from typical gasifiers such as Texaco and KRW do not have the reducing power to form appreciable amounts of CeO_n. Recent research involving three-way automotive and oxidation catalysts has shown that the catalytic performance of CeO_2 is enhanced by the addition of ZrO_2. It is said that, among others, ZrO_2 increases the oxygen reducibility and improves the thermal stability, factors that should also improve the performance of ceria-based desulfurization sorbents in less reducing atmospheres.

CeO_2-ZrO_2, available commercially and synthesized in-house electrochemically are being studied. In-house synthesis provides the flexibility of altering the sorbent structure and composition for optimal sulfur removal. In-house synthesis and characterization of CeO_2-ZrO_2 materials were reported during the 2001 UCR meeting.

Experimental effort during the past year has used a laboratory-scale fixed-bed reactor to determine H_2S removal efficiency as a function of temperature and sorbent and feed gas composition. The reactor and feed and product lines are constructed of quartz, teflon, and silcosteel to eliminate interaction between H_2S and stainless steel surfaces. Product gas concentration is measured by gas chromatography using a pulsed flame photometric detector (PFPD) for low H_2S levels (0.1 to 10 ppmv) and a thermal conductivity detector (TCD) for higher concentrations.

Journal Articles

Electrochemical Synthesis of Nanocrystalline Ceria-Zirconia, A. Mukherjee, D. P. Harrison, and E. J. Podlaha, Electrochemical and Solid State Letters, 4, D5, 2002

Presentations

High Temperature Desulfurization of Synthesis Gas: I., A. Mukherjee, K. B. Yi, E. J. Podlaha, and D. P. Harrison, presented at the DOE University Coal Research Conference, Pittsburgh, June 2001.

Electrodeposition of Ce-Zr Powder, A. Mukherjee, D. P. Harrison and E. J. Podlaha, presented at the Joint International Meeting of the Electrochemical Society, San Francisco, September 2001.

Electrochemical Synthesis and Characterization of $Ce_{1-x}Zr_xO2$ Nanocrystalline Powders, E. J. Podlaha, J. Chan, J. Jiang, A. Mukherjee, and D. P. Harrison, presented at the Annual Meeting AIChE, Reno, November 2001

Ceria-Zirconia High Temperature Desulfurization Sorbents, K. B. Yi, E. J. Podlaha, and D. P. Harrison, presented at the Annual Meeting AIChE, Reno, November 2001.

Students Receiving Support from the Grant

A. Mukherjee (LSU support)
K. B. Yi